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Indoor Air Quality

With 155 Figures and 190 Tables

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Preface

The International Conference on Indoor Air Quality, Tokyo, 1987 was held from November 4–6, 1987, at The New Otani Hotel in Tokyo, Japan, under the auspices of the Council for Environment and Health, whose president is Dr. Hitoshi Kasuga of Tokai University.

The 1980s have witnessed remarkable progress in numerous research programs on indoor air quality. It is noteworthy that the effects of environmental tobacco smoke (ETS) on nonsmokers and of nitrogen dioxide-induced indoor air pollution drew recognition as serious problems not only among epidemiologists, pathologists, and clinicians the world over, but from the general public as well.

There have been significant advances in the area of ETS alone. The separate findings of Takeshi Hirayama and Dimitrios Trichopoulos, released almost simultaneously in 1981, on the relationship between ETS and lung cancer drew immediate attention worldwide and triggered more than 10 follow-up studies. The controversy raised by this work still continues.

A number of international symposiums have been held on this topic, with those in Geneva (1983), Vienna (1984), and Essen (1986) commanding the greatest global attention. Yet, none have established a definite causal relationship between ETS and lung cancer.

A special report in 1986 by the U.S. Surgeon General, entitled "The Health Consequences of Involuntary Smoking", concluded that passive smoking is a cause of disease, including lung cancer, in healthy nonsmokers. This conclusion was reached through exhaustive study and despite many reservations, supporting the findings of Hirayama et al.

At about the same time, the Japanese Ministry of Public Health and Welfare published "Smoking and Health," its first such report, giving mild support to the view that smoking is harmful in stating:

> Although there is currently no worldwide support for the view of there being a significant risk of lung cancer from passive smoking, fear and concern have been expressed over its danger in many countries.

At the annual meetings of the World Health Organization and at the World Conference on Smoking and Health, discussions were based on the assumption of an established link between ETS and lung cancer. These conferences thus provide solid ground for antismoking campaigns.

Dr. Ernest Wynder, a keynote lecturer at the Tokyo conference, touched on one of the grounds for debate on the causal relationship between smoking and lung cancer. He pointed out that since this association is weak,

the conclusions drawn at these conferences are being highly influenced by biases involved in the measured amounts of ETS exposure, questionnaire responses, and subsequent classification of nonsmokers. This perhaps makes evaluation of what Dr. Wynder calls "critical association" extremely difficult.

As one of the planners and organizers of this conference, I believed it possible to objectively and scientifically evaluate this critical association by establishing a clear focus on an issue which has tended to become hopelessly obscured. I thus sought to establish an international forum for researchers to discuss ETS and pool available scientific data on indoor air quality produced over the past several years.

A total of 100 researchers, including younger people in the forefront of research, and leading scholars in their respective fields (60 from abroad and 40 from Japan) were invited to participate in the conference.

The conference opened with keynote lectures by Dr. Ernest L. Wynder, Dr. Barbara S. Hulka, Peter N. Lee, and Hitoshi Kasuga. These were followed by general presentations on ETS Measurement (Sessions 1 to 3), on the Biological Effects of ETS (Sessions 4 to 8), on the Epidemiology of Passive Smoking (Sessions 9 to 11), and on Indoor Air Pollution (Sessions 14 to 17).

Reports from the above presentations were summarized at three panel discussions: Epidemiology of Passive Smoking (Session 12), Reassessment of Passive Smoking as Lung Cancer Risk (Session 13), and ETS Measurement, Biological Effects of ETS, and Indoor Air Pollution (Session 18).

Session 13 proved to be a major highlight of the conference as exciting debate at this evening session extended into late hours.

Some 95% of all those invited attended the conference. Among those unable to come were Dr. Doll, who could not take the trip because of advanced age, and Mr. Garfinkel and Dr. Trichopoulos, who had other academic commitments and sent coresearchers on their behalf.

Special thanks go to Professor G. Lehnert, vice president of the conference, and Professor K. Maeda and Dr. Fukuma, who served as vice presidents and panelists.

I am also very much indebted to Messrs. Y. Yanagisawa and T. Namekata, from the United States, and Professors K. Maeda, K. Aoki, Y. Tsunetoshi, from Japan, who chaired the panel sessions.

Appreciation also is extended to Dr. Shimizu and Dr. Matsuki, who served as secretary general, for their efforts in organizing the conference.

In conclusion, heartfelt thanks go to all conference participants for their cooperation and excellent presentations. I wish them continued good health and success.

November 1989 *Hitoshi Kasuga*
Kanagawa, Japan

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Assessment of ETS Impact on Office Air Quality

J. J. Piadé, C. Gerber, and W. Fink

Summary

The contribution of environmental tobacco smoke (ETS) to indoor air quality was investigated by quantifying the concentration of some of its constituents in the course of a series of strictly controlled experiments.

One brand of commercial cigarettes was smoked by trained smokers following a prescribed protocol both in a test-chamber and in an office of a modern, air-conditioned building. The ETS components investigated were CO, NO, NO_2 and nicotone. The concentration of respirable suspended particles (RSP) was also monitored using three different methods.

The concentrations of these ETS constituents and their ratios are reported, together with background and outdoor levels. In addition, the influence of room ventilation, smoke generation rate, wall deposition effects, etc., is discussed.

Introduction

The indoor air concentration of ETS components has been surveyed by many authors in real-life measurements, but with little or no information on smoke generation. In other reports, mostly for exposure studies, both smoke generation and air concentration of several ETS components were carefully monitored, but with often unrealistic smoke levels [1, 2].

This paper is the first part of a study aimed at investigating ETS chemistry in real-life situations, but with a strictly defined smoke generation and investigating a wide array of components. It comes as a continuation of previous investigations on sidestream smoke (SS) generated in a test-chamber [3]. In this study the effects of smoke generation patterns, room ventilation and air mixing should be assessed, with an emphasis on the time variation of the measured concentrations and their ratios. This paper reports on early results establishing the experimental concept, checking methods and evaluating the impact of various indoor environmental factors.

Experimental Procedures

Smoking Sessions

The office used for this study has a surface of 12 m^2 and a volume of 35 m^3, with a door and a large window. Its walls are plastered, the floor is carpeted and it is furnished with a desk, three chairs and a cupboard. It is situated in a modern building

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with central air conditioning. The ventilation was checked to ensure 3.5 air changes per hour.

Smokers normally consuming about 1 pack per day were trained to take 2-5 puffs per minute in a reproducible way, as checked by consistent puff-counts per cigarette. They were asked to smoke commercial cigarettes according to a pre-determined, realistic protocol. All smokings took place in the same room, but ventilation was turned on or off with possible additional air mixing.

Analytical Methods

For each session, the concentrations of CO, NO, NO_2 and respirable suspended particles (RSP) were measured continuously. Nicotine concentration was measured periodically.

Samplings were done using feed-back flow control pumps (SKC Aircheck Sampler 224-36) drawing air from near the center of the room at an height of about 1.2 m.

Carbon monoxide was measured continuously by non-dispersive IR (Dasibi 3008) and nitrogen oxides by chemiluminescence (Tecan CLD 502).

Nicotine was sampled by pumping air through XAD-4 tubes (SKC 226-30-11-04) which were extracted with 1 ml of ethyl acetate (0.01% triethylamine) and analysed by capillary gas chromatography according to [4]. Quinoline was used as an internal standard.

RSP concentration was simultaneously measured by three different methods:

- Filter gravimetry, by pumping air at 2 l/min through a filter pad (Fluoropore, Millipore FALP03700), possibly after passing through an impactor (TSI 3.5 µ cut-off) retaining particles that would not be inhaled [5], according to [4]. The weight change was measured with a microbalance (Mettler M3).
- Portable piezobalance (TSI model 5500).
- RAM nephelometric detector (GCA RAS-1).

Instrument Calibration for RSP Determination

The gravimetric determination is a direct method which is well established [4, 6]. It is precise down to about 30 $\mu g/m^3$ for 1-h samplings and the coefficient of variation of replicate analyses is about 4%. It only provides time-averaged answers, whereas the RAM gives almost real-time readings and the piezobalance provides a result every 3-5 min.

The TSI 5,500 is factory calibrated and gives direct readings of RSP levels (mg/m^3). It has been used in many ETS studies [7] and its performance has been questioned by several authors [2]. The manufacturer reports it to underestimate tobacco smoke by 15% [9] and in a recent study significant differences between the responses of two identical instruments were reported [8]. The response of the TSI 5,500 we used to SS (between 0.09 and 1.2 mg/m^3) was compared to gravimetric determinations in a series of experiments performed in our test-chamber. The difference between both determinations was consistently smaller than the variability of the methods, provided that the sampling flow rate of the piezobalance was kept at exactly 1 l/min and that its sensor was washed after each determination.

In contrast to the piezobalance, the RAM has to be calibrated before use with the aerosol studied [10]. This is due to its sensitivity to the particle size distribution of the

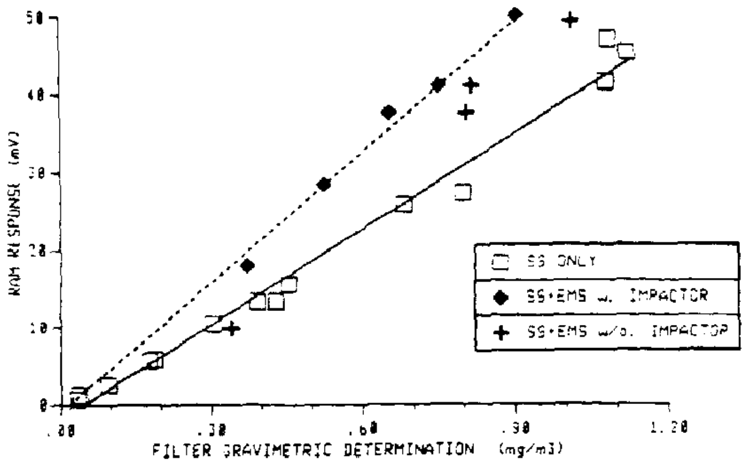

Fig. 1. Calibration of RAM vs. gravimetric determinations

sample. To this aim, the time-averaged RAM output was compared to gravimetric results in a series of experiments where smoke was generated in the test-chamber by SS only (machine smoking, mainstream smoke (MS) exhausted out of the room), or by SS plus exhaled MS (human smoking). Determinations were made for total airborne particulate matter or for RSP only (by sampling through 3.5 μ impactors).

The results are given in Fig. 1. They reveal two possible sources of systematic error:

- If the RAM is calibrated using SS only for ETS measurements, RSP results will be significantly over-estimated.
- It is obvious that omitting the impactor will result in over-estimating the air burden if one should perform a direct gravimetric determination. But since the RAM response is practically not affected by the adjunction of an impactor, it is essential that the calibration be made by comparison with *RSP only* (i.e. using 3.5 μ impactors at the filter and RAM inlets).

Results and Discussion

For each smoking session of this first set of office ETS studies, the smoke generation protocols and the environmental conditions are given in Table 1.

In experiment 3, five cigarettes were smoked simultaneously, and the room ventilation was left on. Time zero was set at the moment when the cigarettes were extinguished. Figure 2 shows the plot, as a function of time, of the CO concentration together with that of RSP as measured with the RAM and with the piezobalance and the time-averaged concentration of nicotine. These values are all background corrected.

Figure 2 shows that the CO concentration decreases exponentially. The calculated decay rate is almost equal to the measured air changes per hour in the room. Thus CO is a good tracer that can be used to offset the effects of room ventilation.

The RSP concentration as measured by the RAM also decreases exponentially, a little faster than the CO. Thus the RSP to CO ratio does not remain constant with time.

Table 1. Smoke generation protocol and environmental conditions

Experiment code	Number of cigarettes smoked	Generation rate	Room ventilation
1	1	at time 0	on
2	2	at time 0	on
3	5	at time 0	on
4	9	every 15 min	on
5	2	at time 0	off
6	4	every 15 min	off
7	4	every 15 min	off, fans on

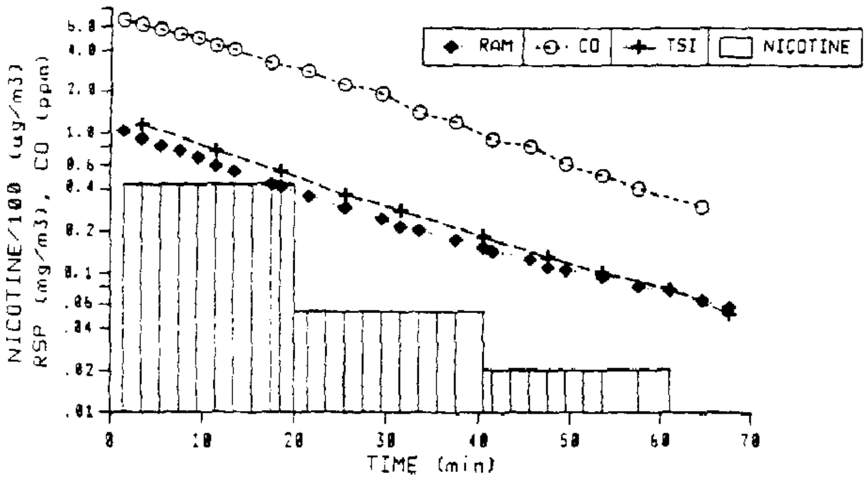

Fig. 2. RSP, CO, and nicotine decay after smoking 5 cigarettes

Actually, the decay rate of the RSP/CO ratio reflects the kinetics of wall impaction and sedimentation of the particles.

If we now consider the piezobalance determinations, they are slightly higher than the RAM measurements for unaged ETS. After about 40 min both curves coincide. An explanation for this discrepancy may be sought in changes in the smoke particle size during the early aging phase [11].

The plot of the nicotine concentration shows that it decays much faster than RSP immediatly after smoking. After 1 h, the level drops much more slowly, actually even more slowly than the CO. This is probably due to the fact that nicotine is mostly present in the gas phase [12], and wall effects become very important. Of course the nicotine/RSP ratio is far from remaining constant.

Figure 3 shows the time variation of NO and NO_2 concentrations. The decay of the NO concentration appears to be exponential. Considering the NO/CO ratio, which offsets the effect of room ventilation, evidences the contribution of what seems to be a pseudo-

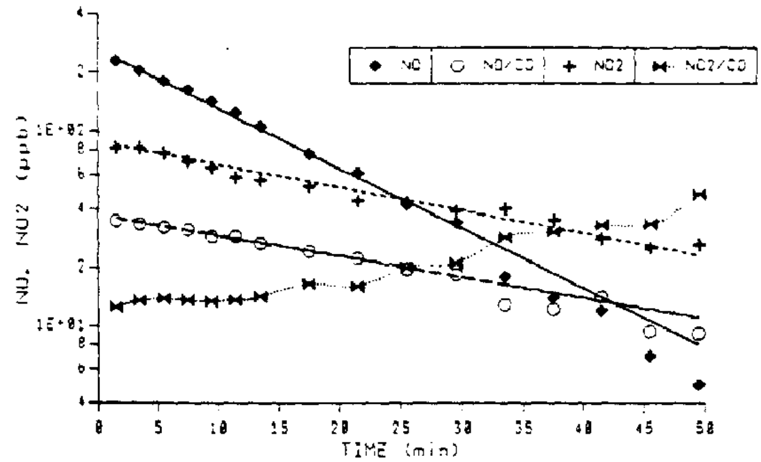

Fig. 3. NO, NO/CO, NO_2, and NO_2/CO decay after smoking 5 cigarettes

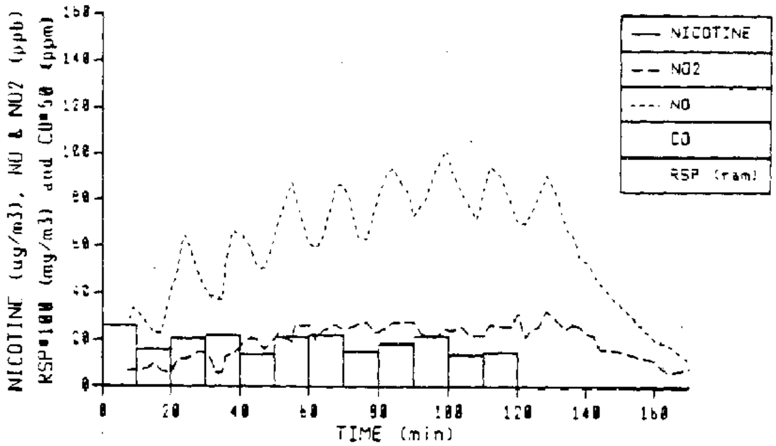

Fig. 4. RSP, CO, NO, NO_2, and nicotine concentration; 9 cigarettes smoked at 15-min-intervals

first order chemical decay. It should be noted that the NO decay was recently reported to be pseudo-first order in MS gas phase, but pseudo-second order in the whole MS [13]. The time increase of the NO_2/CO ratio, on the other hand, reveals a chemical generation of NO_2 in the early phase of ETS aging. Of course, the NO_2 level decreases in absolute value after a few minutes.

A steady-state situation can be created with a constant smoke generation rate. This is what is obtained in experiment 4, where a cigarette is smoked every 15 min with the room ventilation left on. The corresponding profiles are shown on Fig. 4.

Table 2. Time averaged RSP and nicotine concentrations

Experiment code	RSP		Nicotine	
	Sampling time (min)	Concentration (mg/m^3)	Sampling time (min)	Concentration ($\mu g/m^3$)
1	90	0.089	40	8.6
2	96	0.189	40	18.6
3	79	0.391	40	25.6
4	150	0.478	40	21.4
5	121	0.350	40	25.3
6	128	0.508	40	28.7
7	130	0.486	40	16.8
Indoor background		0.033		0.7

Each time a cigarette is smoked, there is a rise and subsequent decay of the CO, NO and RSP concentrations, and after about 1 h a steady-state concentration is achieved. Even the nicotine level becomes fairly constant after a brief initial peak. This kind of experiment could be very useful in determining how environmental conditions may affect the ratio between the concentrations of two ETS components.

The effect of changes in the environmental conditions can also be quantitatively evaluated when the time-averaged nicotine and gravimetric RSP concentrations obtained for all the situations investigated are compared. These results are gathered in Table 2 and perusal of this table allows the following comments to be made:

Comparing the RSP and nicotine averaged concentrations in experiments 1, 2 and 3, it appears that these values are not proportional to the number of cigarettes smoked, even in this strictly controlled set of experiments. This is even more true for the nicotine values and thus the nicotine to RSP ratio is fairly different in these three experiments. The drastic effect of room ventilation is obvious when comparing the results of experiments 2 and 5 or, in the case of continuous smoke generation, 4 and 6. Again, the impact of room ventilation is quite different whether one considers RSP or nicotine. Eventually, the effect of an increased air turbulence in the room is apparent when comparing the results of experiments 6 and 7. It appears that the average concentration of nicotine is much more reduced by air turbulence than that of RSP, pointing at the large influence of wall effects on nicotine concentration.

Background Indoor and Outdoor Levels

In average, the indoor background levels were about 0.6 ppm for CO, 10 ppb for NO, 50 ppb for NO_2, 30 $\mu g/m^3$ for RSP and 0.7 $\mu g/m^3$ for nicotine

In addition to indoor analyses, and in order to put these results in perspective, the outdoor concentration of CO, NO and NO_2 was measured, at the same time as the smoking sessions were held, by extending probes 1 m outside the window. The levels monitored over a 24-h period are plotted on Fig. 5. For nitrogen oxides, these values are at times higher than any level obtained in the course of our experiments. This is due in part to the proximity of a highway.

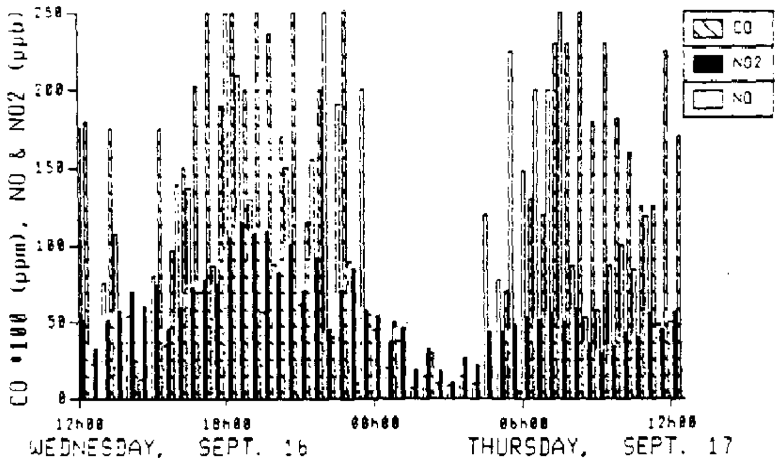

Fig. 5. Outdoor CO, NO, and NO_2 concentrations

Conclusion

This study constitutes a first part of a program we have initiated on the analytical investigation of ETS in indoor air. Much more work is needed to obtain a good understanding of the main processes governing ETS aging. This initial study outlined some possible flaws in RSP measurement. It showed that a careful examination of the time variation of the measured concentrations and their ratios may yield valuable insights into ETS aging processes. As these ratios are not constant, it appears that no component can readily serve as a marker for other ETS components. In particular, nicotine was found to be quite outstanding in its behaviour, making it a poor marker of ETS exposure. Eventually the large impact of indoor environment factors such as air mixing, room ventilation, wall surfaces etc. on ETS was outlined.

References

1. National Research Council (1986) The physicochemical nature of sidestream smoke and environmental tobacco smoke, measuring exposures and assessing health effects. National Academy Press, Washington DC, pp 25–53
2. Sterling TD, Dimich H, Kobayashi D (1982) Indoor by-product levels of tobacco smoke, a critical review of the literature. JAPCA 32:250–259
3. Blake CJ, Piadé JJ, Fink W (1986) Quantitative evaluation of cigarette sidestream smoke components under controlled experimental conditions. Presented at the International Experimental Toxicology Symposium on Passive Smoking, Oct. 23–25, 1986, Essen (FRG)
4. Ogden MW, Conner JM (1987) Methods of analysis for nicotine, respirable suspended particles and ultraviolet particulate matter in environmntal tobacco smoke. Collaborative Study; Presented at the 41 Tobacco Chemists' Research Conference, Oct 4–7, 1987, Greenboro NC (USA)
5. Heyder J (1985) Studies of particle deposition and clearance in humans. In: Grosdanoff P et al (eds) Problems of inhalatory toxicity studies. BGA Publ, MMV-Medizin Verlag, Munich

6. Pio CA (1986) General sampling techniques. In: Harrison RH et al (eds) Handbook of air pollution analysis. Chapman and Hall, London New York
7. Repace JL, Lowrey AH (1983) Indoor air pollution, tobacco smoke and public health, part I; Science 208 (1980) 464–472, Part II; World Smoking and Health 7(4) (1983) 35–46
8. Ingebrethsen BJ, Heavner DL, Angel AL, Conner JM, Oldaker GB, Green CR (1986) A comparative study of environmental tobacco smoke particulate mass measurements in an environmental chamber. Presented at the International Experimental Toxicology Symposium on Passive Smoking, Oct. 23–25 1986, Essen (FRG)
9. Sem GJ, Tsurubayashi K, Homma K (1977) Performance of the piezoelectric microbalance respirable aerosol sensor. Am Ind Hyg Assoc J 38:580–588
10. Rawbone RG, Burns W, Patrick RA (1987) The measurement of environmental tobacco smoke particulates. Toxicology Lett 35:125–129
11. Ingebrethsen BJ (1986) Aerosol studies of cigarette smoke. Rec Adv Tob Sci 12:54–142
12. Eudy LW, Thome FA, Heavner DL, Green CR, Ingebrethsen BJ (1985) Studies on the vapor/particulate phase distribution of environmental nicotine by selective trapping and detection methods. Presented at the 39 Tobacco Chemists' Research Conference Oct. 2–5 1985, Montreal (CAN)
13. Borland CDR, Chamberlain AT, Higenbottam TW, Barber RW, Thrush BA (1985) A comparison between the rates of reaction of nitric oxide in the gas phase and in whole cigarette smoke. Beitr Tabakforschung Int 13(2):67–73